LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

LLNL-PROC-426088

Radiochemistry diagnostics for the National Ignition Facility

R. D. Hoffman, C. J. Cerjan, D. A. Shaughnessy, K. J. Moody, S. L. Nelson, L. A. Bernstein, M. A. Stoyer, R. J. Fortner, D. H. Schneider

March 18, 2010

Nuclear Explosives Design Physics Conference
livermore, CA, United States
October 26, 2009 through October 30, 2009

Disclaimer

Radiochemistry diagnostics for the National Ignition Facility (U)

R. D. Hoffman, C. J. Cerjan, D. A. Shaughnessy, K. J. Moody, S. L. Nelson, L. A. Bernstein, M. A. Stoyer, R. J. Fortner, and D. H. Schneider

Lawrence Livermore National Laboratory, Livermore, CA

Radiochemistry-based techniques will be an important complement to x-ray implosion diagnostics. Simulations demonstrate the value of alpha-induced or deuteron-induced reactions as a direct measurement of static mix contamination in DT-filled ignition capsules. In this proceedings we examine to what extent neutron-induced reactions might be highly correlated with the energetically down-scattered neutron fraction which is, in turn, related to the critical quantity of fuel areal density, ρR. Although alpha and deuteron reactions are highly suppressed in HT/D-filled capsules, neutron-induced reactions produce robust abundances and 2% measurements of the relevant radiological ratios is achievable. We conclude that radiochemical data are strongly correlated with the down-scattered fraction and fuel areal density. Unknown physics, primarily uncertainties in the direct T-T fusion neutron cross section and, to a lesser extent, various radiochemical production cross sections are important but calibration shots can be used to reduce these errors. The primary advantage of radiochemistry remains -- it is the only technique that samples down-scattered neutrons from the entire capsule during the burn. In particular radiochemistry correlated with other diagnostics can be used to minimize experimental uncertainties and thus maximize gain. (UNC).

Motivation

Radiochemistry implosion diagnostics have the advantage of being inherently timed to the nuclear burn; they are not limited to a particular line-of-sight, are insensitive EMP; and do not require precise timing alignment with peak burn. In addition, appropriate choices of reaction products may simultaneously diagnose several important implosion characteristics. In particular, charged particle reactions are sensitive to hot spot conditions while neutron-induced reactions are sensitive to the dense, colder surrounding material. Indeed, simulations using DT-filled capsules have demonstrated that alpha-induced reactions directly measure fill tube material injected into the ignition hot spot and that neutron-induced reactions are highly correlated with the energetically down-scattered neutron fraction[1,2] .

As a precursor to a high yield ignition attempt, hydrodynamic-equivalent, low yield targets will be fielded. The only difference between the high- and low-yield ignition attempts will be the gas fill and cryogenic layer, which will be approximately 3:1 tritium to hydrogen with variable amounts of deuterium. Implosion experiments using HT/D-filled capsules are less straightforward to analyze using radiochemical signatures when compared to their DT-filled counterparts. The subject of this paper is to discuss these differences. The T + T -> α + 2n fusion reaction (TT), is such a prolific neutron source that the neutron-induced reactions that probe fuel areal density produce easily detectable abundance levels. Diagnostic complication arises from details of the statistical spectrum of the emitted neutrons from the TT fusion reaction, which span an energy range from 1 to 10 MeV. This energy range also contains down-scattered neutrons from the residual DT-fusion reaction so that the recovered product abundance includes contributions from both the direct and down-scattered neutrons. Since the radiochemical techniques sample all available neutrons, effort must be taken to understand the relative contributions of the signal that are sensitive to the fuel areal density from those that are not.

Further evaluation of the radiochemical areal density signature arise from the uncertainty in the reaction cross sections in any of the considered neutron-induced reactions such as (n,2n) and (n,γ). Thus any

abundance measurement is not an absolute measurement but instead will require calibration to establish its specific value. Calibration issues are less important when comparing differences between nearly similar shots. Another issue, which we will address, resides in potentially subtle differences in late-time dynamics of the implosion process. For example, (n, γ) reactions have cross sections that rise as a function of decreasing energy so that differences in the late-time contributions in the implosion dynamics , particularly for HT/D shots, are important in comparison of radiochemical signatures.

These topics will be addressed below in the specific context of a fuel areal density diagnostic for TH/D-filled capsules. More precisely, attention will be focused on the viability of a particular set of neutron-induced reactions: $^{124}Xe(n,2n)^{123}Xe$ and $^{124}Xe(n,\gamma)^{125}Xe$. Determining the ratio of the two products should be experimentally straightforward and reliable to sufficient accuracy. So the primary issue remains the relationship of the measured quantity to the fuel mass distribution under the likely implosion conditions of the HT/D shot campaign.

Technical Details

Downscattered Spectrum

A typical escaped neutron energy spectrum is shown in Figure 1 (top panel) for an HT/D capsule fill (0.7485 T, 0.2495 H, 0.002 D) with (red curve) and without (green curve) elastic neutron scattering processes. The salient differences in the scattering contributions are clear in the range 9-13 MeV and below 2 MeV. Of course scattering processes shift the neutron energy over the entire range but this plot emphasizes the energetic downscattering of the direct 14 MeV DT neutrons most visible in the 10 – 12 MeV range and the analogous downscattering from the TT direct reaction below 2 MeV. The energy range from 2 – 9 MeV includes both direct and energetically downscattered contributions from both DT and TT. Also shown are two relevant energy ranges for D-T (direct 12-17 MeV and energetic downscatter 10-12 MeV) sampled by an ideal 4π neutron time of flight diagnostic.

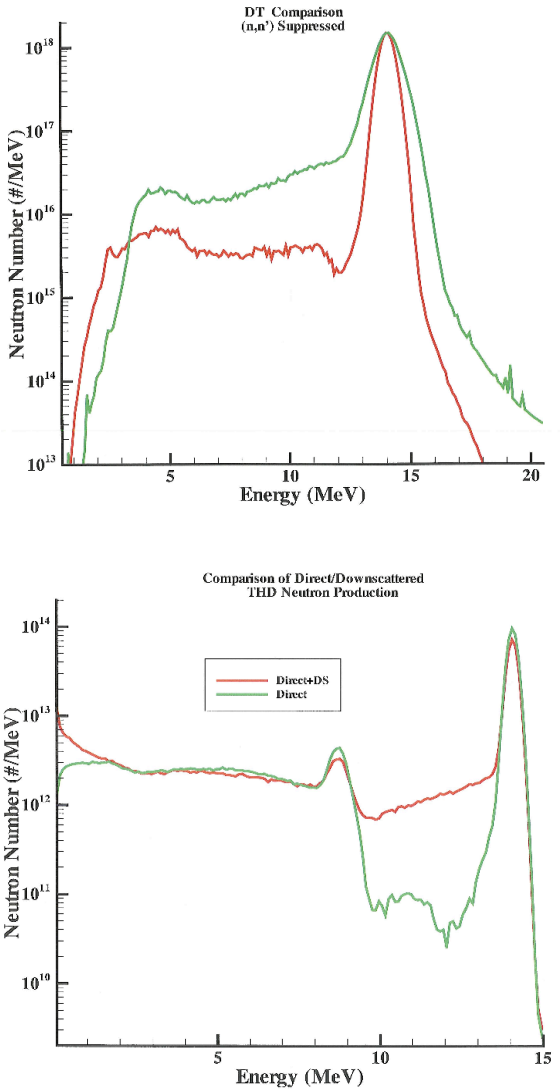

Fig. 1. Escaped neutron spectrum for DT (top panel) and HT/D (bottom panel) gas fills showing the direct and downscattered contributions as simulated with and without elastic neutron scattering processes.

An analogous comparison for equi-molar DT-gas filled capsules is also presented in Figure 1(bottom panel). The energy region between 3 and 12 MeV likewise displays the primary difference in the inelastic scattering contributions.

It is noteworthy that the escaped neutron spectra differ qualitatively due to the diminished (TT) fusion reaction contribution and to the much more rapid disassembly of the high yield capsule.

Cross Sections

Select neutron-induced reactions can discriminate various spectral features. Although many possible dopants have been investigated, those that have noble gas products will be the first ones employed since gas collection should be logistically simpler than solid debris collection. We consider the element xenon to be an ideal first choice with many potential advantages. It has 9 stable isotopes and many attractive activation products with suitable half-lives (hours-days), including ones that are shielded from potential contamination from fission-fragment decay. Loading separated isotopes (including radioactive ones) allows for a potential collection efficiency determination. Previous experience with noble gas collection in ICF environments has demonstrated up to 95% collection efficiency[3]. Typical loading of ~10^{14} atoms easily provides the 10^6 – 10^8 atoms needed for a 2-5% measurement error. Finally, the necessary cross sections have been recently evaluated[4].

In Figure 2 we show the ^{124}Xe (n, 2n) ^{123}Xe and the ^{124}Xe (n, γ) ^{125}Xe cross sections as a function of incident neutron energy. The (n,2n) reaction preferentially reacts with the primary (14 MeV) neutrons produced in the DT fusion reaction, whereas the (n, γ) reaction is primarily sensitive to neutrons with energy below 4 MeV. Although the separate abundances provide information about the neutron energy spectrum, their ratio [(n, γ)/ (n,2n) , hereafter referred to as 125/123] yields a more easily interpreted value since the effects of the capsule shell's non-uniform spatial distribution and differences in absolute neutron number (yield) are minimized. Also shown for both reactions is a comparison to experimental data.

For ^{124}Xe (n, γ) ^{125}Xe the measured energy range is between 5 to 100 keV with a scatter of 12%. For ^{124}Xe (n,2n) ^{123}Xe the experimental error is 3%, but data is only available at 14 MeV. These are typical uncertainties for (n,γ) and (n,2n) reactions on the other stable xenon isotopes, and our calculated cross sections typically agree within 10% for (n,2n) and 20-40% for (n, γ).

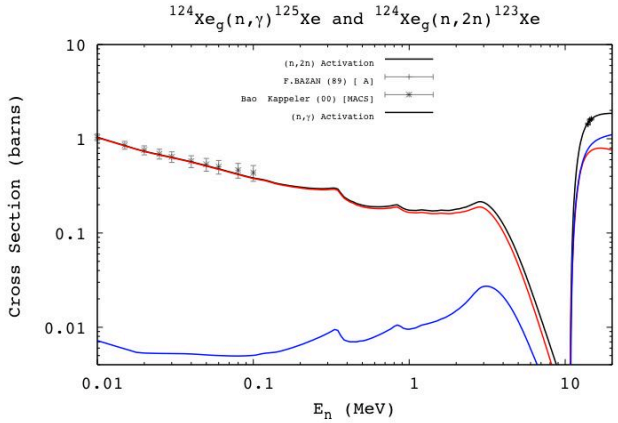

Fig. 2. ^{124}Xe (n,2n) and (n,γ) reactions for a fuel areal density diagnostic compared to experimental data[5,6].

It should be noted that the low energy cross section for the (n, γ) reaction rises in the very low energy regime. This dependence generally holds for this class of reaction so that lower energy neutrons may contribute a substantial fraction to the overall production. As will be discussed below, the low energy contribution may constitute more than half of the observed product abundance.

Based on figures 1 and 2 we can make some qualitative statements immediately. The DT case is straightforward, with the (n,2n) signal caused primarily by 14 MeV neutrons, whereas the (n, γ) signal is caused by downscattered neutrons and thus measures the areal density(ρR). Direct contributions from TT neutrons are small and the short confinement time minimizes contributions from low energy neutrons. For the HT/D case the situation is more complicated. Here the (n,2n) signal is strongly suppressed, while the (n, γ) signal has two contributions, a direct contribution from primary TT neutrons and a downscattered component mostly from contributions below 2 MeV. As we will see in our analysis below most of the radiochemical signal comes from the region below 2 MeV.

Radchem Ratio Correlation to nTOF

In any experimental campaign the first measurements are expected to be from capsules with the relevant amounts of H, D, and T, but designed to have a low areal density (exploding pusher) so that the radiological signal associated with the direct neutrons (green curve in Fig. 1) could be measured and thus reduce experimental uncertainty. It should be emphasized that the primary difference in the radiological signal between HT/D and DT is that longer confinement time and increased lower energy scattering from TT neutrons produce an additional source of signal from neutrons below 2 MeV but this signal is remains proportional to the fuel areal density.

Figure 3 shows a comparison of our radiochemical signature with an ideal neutron time of flight (nTOF) diagnostic that would be capable of looking at all angles. A strong correlation exists between the (n,γ)/(n,2n) ratio and downscattered fraction, as might be expected on physical grounds. In the top panel this correlation is shown for a set of DT-filled capsule simulations, where the relevant nTOF fraction is the ratio of the 6-10 MeV to the 12-17 MeV signal. In the center panel we show the correlation of the 125/123 product ratio for some HT/D(0.005)-filled capsule simulations, where the nTOF ratio is now the 10-12 MeV to 12-17 MeV signals. Note that this ratio will depend upon the capsule configuration -- most importantly the amount of D in the HT/D-filled capsules. In the bottom panel we plot both of these correlations vs. ρR.
It should be emphasized that our comparisons with nTOF are the, so-called, 4π nTOF.

The radiochemical signature is insensitive to mass distribution and provides an independent verification of the reliability and possible issues of anisotropy of the nTOF-determined fraction.

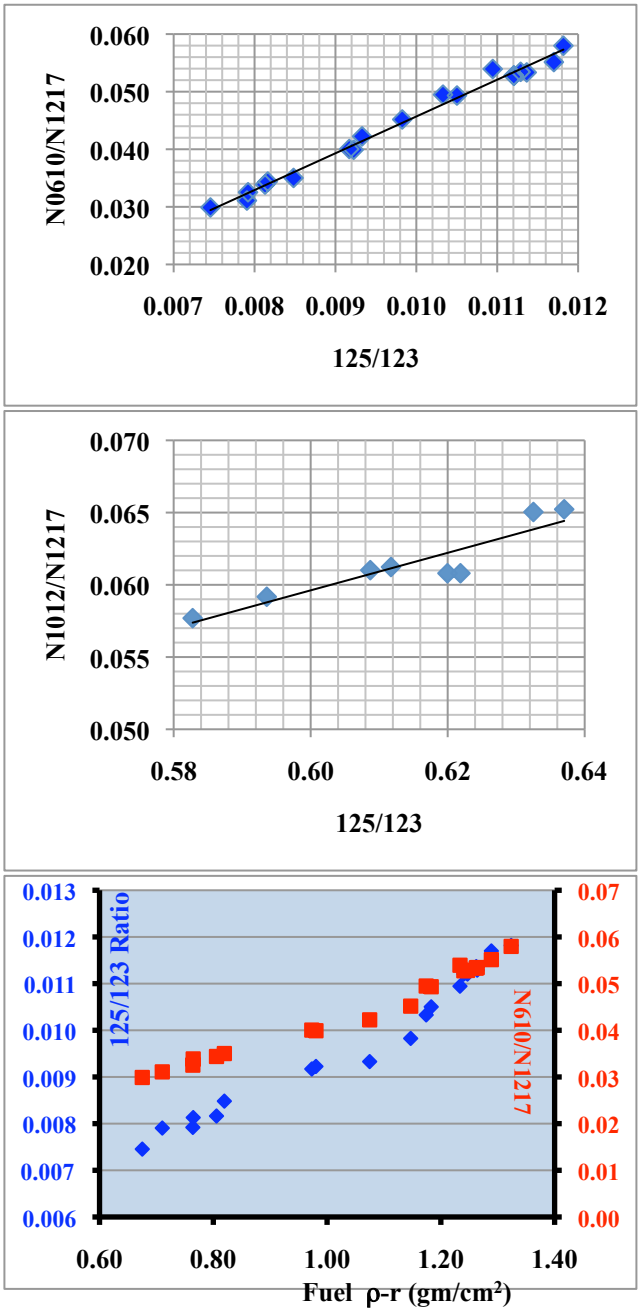

Fig. 3. Correlation between the 125/123 ratio and downscattered neutron fraction for equi-molar DT (top) and HT/D (center) simulations. The bottom panel plots both as a function of fuel ρR.

Note that the (125/123) product ratio may be generally understood as the ratio of the sum of the low energy direct neutrons plus the lower energy downscattered neutrons divided by the primary neutron number: (D + DS)/P. In the limit of negligible downscattered neutron contribution, the nTOF fraction will vanish whereas the (125/123) ratio will not. It is expected that calibration shots using low ρR targets would be used to minimize the fixed "background" signal for this series of experiments.

Temporal Dependence

Finally we consider the temporal dependence and the range of neutron energies that contribute to the xenon isotope 125/123 ratio. Based on what we have seen, we expect the (n,2n) signal to follow the burn; we also expect the time dependence of the signal to persist later in time; and we expect the contributions from lower energy neutrons in the HT/D shots to be much larger.

In Figure 4 we compare the individual ^{123}Xe and ^{125}Xe production rates and their product ratio [125/123] vs. offset from peak burn time for an equi-molar DT and an HT/D(0.005) simulation with a very low yield. As a reference, energy production is also plotted (black curve). The DT signals (upper panel) are not surprising. The ratio (green curve) follows the burn closely. For the HT/D shot (lower panel) the behavior is similar: it takes longer to burn, peak ^{125}Xe production occurs past peak burn, and there is a pronounced weak tail in the (n,γ) signal late in time.

Clear differences in the temporal dependence of individual radiochemical species for the DT-filled and HT/D-filled capsules reflect the relative importance of the downscattered neutron contribution compared with the direct TT fusion reaction contribution. An intuitive picture for each of the radiochemical diagnostic products is clear from this plot. The ^{123}Xe product (from the (n,2n) reaction) is almost entirely driven by the high energy 14 MeV neutrons (see Fig. 1); the ^{125}Xe product (from the (n,γ) reaction) primarily tracks the immediate (primary) downscattered neutrons but also has

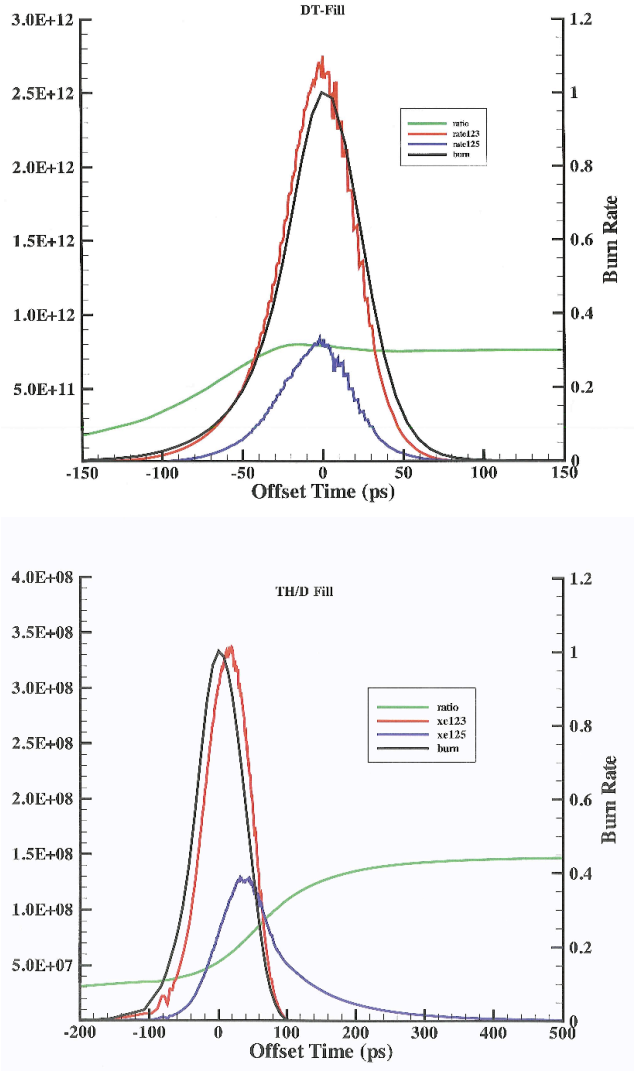

Fig. 4. Xenon isotope production and the 125/123 ratio vs. offset time from peak burn for DT (upper) and HT/D (lower) capsule simulations.

contributions from the statistically distributed TT fusion neutrons and, at later time, lower energy downscattered neutrons. Note that the ratio has achieved 95% of its final value at 350 ps past peak burn time, at the point where the TT reactions are complete. This provides a rough estimate of the

expected error (5%) in this ratio due to uncertainties in the low energy range of the neutron capture cross section. Also notice that the uncertainty due to the TT reactions is absent in the DT capsule burn.

Energy Dependence

For the same two DT and HT/D simulations just discussed we plot in Figure 5 the time-integrated production of ^{125}Xe due to different incident neutron energies vs. offset from peak burn time. Four discrete ranges were chosen to highlight the late time low energy contributions in Fig. 1: E_0 (0 – 200 keV, red), E_1 (200 keV – 1.5 MeV, green), E_2 (1.5 MeV – 11.5 MeV, blue) and E_3 (11.5 MeV – 35 MeV, magenta). The relevance of these energy bins is that for DT shots the three lowest energy bins all contribute to the downscattered fraction from primary 14 MeV neutrons. For HT/D shots, the lowest two energy bins are populated by downscattered neutrons; the intermediate bin E_2 is the sum of downscattered and primary TT neutrons.

For equi-molar DT-filled capsules, the contributions from the lowest three energy ranges (E_0 – E_2) are nearly equal at all times and persist for approximately the same time span. The time-integrated values reflect this balance, which is 0.33 for each of the three lowest energy bins. There is a long-lived component associated the lowest energy bin but it contributes less than 1% to the total production.

The time dependence for TH/D-filled capsules is qualitatively different due to the prominence of the TT fusion reaction neutrons and their downscattered component. In this instance, the lowest energy bin (E_0) dominates the production and has an enhanced persistence in time. When the abundances are integrated over time, the two lowest energy bins contribute about 84% of the total abundance. The uncertainty caused by the direct TT neutrons is contained in the remaining 16%. This is expected to be a fixed quantity relative to the (n,2n) contribution and could be quantified with calibration shots effect on the total, time-integrated abundance.

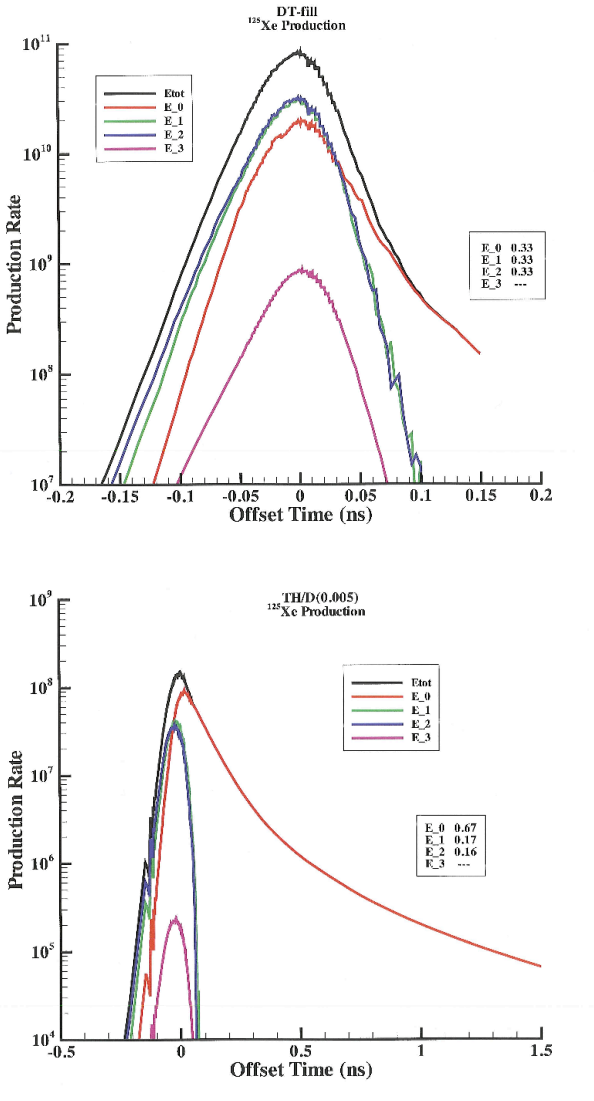

Fig. 5. Contribution of ^{125}Xe isotope production from specific ranges of incident neutron energy vs. offset time from peak burn for DT (upper) and HT/D (lower) capsule simulations.

Radiochemical Gas Sampling

Radiochemical analysis of gaseous samples (RAGS) is a diagnostic instrument designed to collect noble gas products present in the NIF chamber following a shot. Gases will be pumped out of the chamber into the RAGS apparatus, which will then use a series of

cryo-collection stations to isolate and purify noble gases (Figure 6). Implementation of the instrument will focus initially on the collection of xenon gas with additional noble gases added during later campaigns.

The stable tracer ^{124}Xe will be added to the innermost layers of the NIF capsule ablator and will undergo neutron activation reactions during a shot. After passing through RAGS, the collected xenon isotopes will be counted with γ-radiation detectors. As discussed above, ^{124}Xe will undergo (n,γ)^{125}Xe and (n,2n)^{123}Xe reactions. The ratio of ^{125}Xe/^{123}Xe will be reported following a shot with 2% measurement uncertainty, which includes measurement statistics and uncertainty in detector calibration.

The measured isotope ratio will be subsequently interpreted by NIF capsule designers using physics codes as described in previous sections. We estimate approximately 20% absolute uncertainty. This includes uncertainties due to modeled cross sections of the xenon neutron reactions (18%), contribution from TT neutrons (5%), and physics codes (currently 7%). The previously mentioned calibration shots, consisting of exploding pusher capsules with xenon added to the fill gas, will provide baseline values for the contributions of TT neutrons in the limiting case of low ρR capsules. An evaluation of the TT neutron spectrum, particularly at low energies, would remove the 5% uncertainty associated with contributions from direct TT neutrons to the ^{125}Xe value. An experimental effort aimed at measuring neutron-induced cross sections on xenon isotopes, particularly ^{124}Xe, would also lower the absolute uncertainty associated with interpretation of data by reducing the errors on neutron cross sections from 18% down to roughly a few percent.

Uncertainties related to how the physics codes subsequently use and interpret radiochemical data cannot be addressed through calibration or separate measurements and represent a lower limit on the magnitude of the absolute uncertainty associated with the radiochemistry diagnostic. But it is expected that 2% changes in the xenon isotope ratio from shot-to-shot can be detected.

Measurement and Calibration Uncertainties.

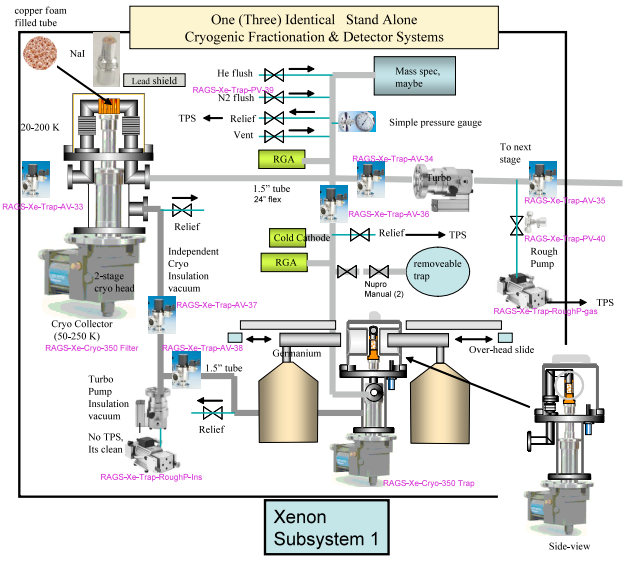

Fig. 6. Schematic of the RAGS noble gas sampling system.

The statistical uncertainty on a measurement of the ^{125}Xe/^{123}Xe isotope ratio is based on a simulated xenon γ-ray spectrum from a high-yield ignition capsule. This includes an assumed Compton background in the detector from fission products present in the samples from a uranium hohlraum. Delaying sample counting by a few hours improves the signal-to-fission background. Simulated γ-ray spectra indicate that even with a fission background present in the detectors, the xenon isotopes of interest should be present in amounts at least 10 times higher than the corresponding fission background.

Lower yield capsules produce fewer activation counts, but count times can be increased to compensate. For example, a simulated HT/D capsule (0.5% D) with 2.6x10^{13} atoms of ^{124}Xe loaded in the inner-most region of the ablator with a yield of 800 J would produce approximately 2.5x10^{7} atoms of ^{125}Xe and ^{123}Xe. Assuming a 100% xenon collection yield (appropriate for noble gas collection), a standard 10% efficiency germanium γ-ray detector, and a delay before counting of two hours (to allow short-lived fission products to decay away), on order of 10^{6} atoms of each of the xenon isotopes would need to be

produced for a measurement with 2% precision. Even for the low yield case, more xenon atoms would be created than required by an order of magnitude. The estimate of 2% total measurement accuracy is currently applied to a range of capsule yields. Conservative estimates were used for RAGS pumping efficiency, collection time, counting delay, count time, detector efficiency, and counting statistics (both signal and fission background). These estimates were based on previously measured xenon spectra and the well-known behavior of detectors at LLNL. It was assumed that only xenon isotopes were present during counting and other nuclides were eliminated through RAGS prior to counting. The 2% uncertainty is valid only for the reported 125/123 ratio (not absolute isotope concentrations) based on experience counting radionuclides.

The (n,γ) and (n,2n) cross sections for ^{124}Xe have not been measured over the entire energy range where reactions will occur in a NIF capsule. Figure 2 shows the modeled cross sections with the data points that are currently available. Incorporating the errors on the measurements (12% for (n,γ) over a neutron energy range of 5-100 keV and 3% for (n,2n) at 14 MeV) and uncertainties in the remaining extrapolated cross section, an uncertainty of 18% is applied to the ratio of the neutron cross sections over the NIF neutron spectrum. An experimental effort to measure these cross sections over a larger energy range than currently reported could potentially reduce this 18% uncertainty and improve the overall absolute uncertainty of a given radiochemical measurement. This value will remain constant for all capsule fills and will cancel out in relative shot-to-shot measurements. The same calibration shots described above can also be used to better quantify these cross sections and set a baseline value that can be applied to subsequent shots.

Conclusions

Radiochemistry techniques will provide valuable diagnostic information about capsule performance during the critical burning phase. The discussion above has concentrated on the near-term application of neutron-activated products and inert gas collection with the limited goal of fuel areal density determination. Despite the complications present at low deuterium atomic weight fraction, relative measurements will be useful. At higher concentrations, these measurements become increasingly unambiguous and possibly quantitative.

Perhaps the most important radiochemical diagnostic will be the determination of cold fuel or shell material mixing into the hot fill gas. It appears that radiochemistry might be the only viable probe of mixed material. Previous work focusing on charged particle reactions (alpha- and deuteron-induced) in a double shell capsule design with loadings similar to those studied here for Xe suggested production of noble gas products (^{21}Ne and ^{79}Kr) in amounts that could be readily collected[7]. Hence, future research will focus on this application.

Acknowledgments

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

References

1. “Ignition failure mode radiochemical diagnostics: initial assessment”, R. Fortner, L. Bernstein, C. Cerjan, *et al.*, LLNL-TR-230178 (2003)

2. “Radiochemistry: A versatile diagnostic for the NIF ignition campaign”, M.A. Stoyer, C.J. Cerjan, K.J. Moody, R.D. Hoffman, L.A. Bernstein, D.A. Shaughnessy, UCRL-TR-405043 (2008)

3. “Planning for Radiochemistry Experiments at the National Ignition Facility (NIF)”, M. Stoyer, UCRL-PRES-214832 (2005)

4. “Neutron and charged-particle induced cross sections for radiochemistry in the region of iodine and xenon”, R. Hoffman, F. Dietrich, R. Bauer, K. Kelley, & M. Mustafa, UCRL-TR-206721 (2004)

5. “Neutron corss sections for nucleosynthesis studies”, Z.Y. Bao *et al.*, Atomic Data & Nuclear Data Tables 76, 70 (2000)

6. “(N,2N) cross-section measurements of krypton and xenon isotopes” F. Bazan, LLNL Nucl. Chem. Ann. Rpt. UCAR-10062-A8 pg 3 (1988)